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Doniach diagram for ordered, disordered and underscreened Kondo lattices.

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The Doniach's diagram has been originally proposed to describe the competition between the local Kondo effect and the intersite RKKY interactions in cerium compounds. Here we discuss the extension of this diagram to different variations of Kondo lattice model. We consider a) ordered cerium compounds where the competition between magnetic order and Kondo effect plays an important role, as in $CeRh_2Si_2$, b) disordered cerium systems with competing spin glass phase, magnetic ordered phases and a Kondo phase, as in the heavy fermion cerium alloy $CeCu_xNi_{1-x}$ and, c) uranium compounds where a coexistence between Kondo effect and ferromagnetic order has been observed, as in UTe. We show that all these cases can be described by a generalized Doniach phase diagram.

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I. INTRODUCTION

The magnetism in strongly correlated f-electron systems is a subject of great interest from both experimental and theoretical points of view, since the famous explanation of the logarithmic decrease of the electrical resistivity in magnetic dilute alloys – like AuFe – by J. Kondo in 1964[1]. This phenomenon has been called Kondo effect, and in the following forty years it has been experimentally observed in many heavy fermion compounds with cerium, ytterbium, uranium and other rare earth or actinide elements. An exact solution of the single impurity Kondo effect has been theoretically determined by the renormalization group and by Bethe ansatz: in essence, the Kondo effect occurs because the localized magnetic moment $S_f = 1/2$ of the magnetic impurity is completely screened by the conduction electron spin-density "cloud" at $T = 0$. For a review see Ref .[2,3]. However, the very rich phase diagram of dense heavy fermion compounds can not be explained in the framework of the one impurity model. It has been proven that the Kondo lattice (KL) model is the appropriate tool for describing the properties of many cerium and uranium compounds.

In heavy fermion materials there are two kinds of electrons: conduction electrons from outer atomic orbitals, and strongly correlated electrons from inner f-orbitals, the later ones being generally localized. As a consequence, the f-electrons may be considered as a lattice of localized spins and the KL model describes the coupling of these localized spins with the electrons in the conduction band. Historically, the KL model has been proposed to account for properties of cerium compounds[4,5]. In most of cerium-based compounds, cerium ions are in the $4f^1$ configuration. In this case, there exists one f-electron per site, with spin $S = 1/2$ spin, which couples antiferromagnetically to the conduction electron spin density, via an on-site exchange interaction J_K. Besides, the local coupling between $f-$spins and conduction electrons may give rise to magnetic order through the RKKY interaction. The competition between Kondo effect, which is characterized by an energy scale $T_K \sim exp(-1/J_K)$, and magnetic order, characterized by the strength of the RKKY interaction $T_{RKKY} \sim J_K^2$, leads to a very rich phase diagram with possible quantum phase transitions. In cerium compounds the transitions typically occur between non-magnetic (Kondo) and antiferromagnetically ordered metallic states. For small values of J_K coupling, the RKKY interaction is dominant and the system orders magnetically. At intermediate values of J_K, energy scales T_K and T_{RKKY} are of comparable strength – magnetic order still occurs but with partially screened localized moments. With further increase of the J_K coupling, the magnetic order is further suppressed and only short range magnetic correlations can survive[4,5] .

The presence of disorder in alloys can strongly affect the competition between the RKKY interaction and the Kondo effect. The role of disorder can be also studied in the framework of the KL model if one considers the interaction between localized spins as random variables with zero mean. This extension is called disordered Kondo lattice (DKL) model. In the DKL model disorder produces a broad distribution of Kondo temperatures and can be responsible for the deviation from the Fermi liquid behavior found in some heavy fermion compounds. Indeed, various effects of "Kondo disorder" has been reported in both cerium based – $CeNi_{1-x}Cu_x$[6,7,8], $Ce_2Au_{1-x}Co_x$ Si_3[9]–, or uranium based – $UCu_{5-x}Pd_x$[10], $U_{1-x}La_xPd_2Al_3$[11] – compounds. Moreover, most of these compounds exhibit coexistence of spin glass and Kondo effect.

Finally, let us specially discuss the behavior of uranium compounds, which is generally quite different from cerium compounds, particularly due to the existence of several coexistence phenomena be-

tween magnetism, Kondo effect and superconductivity[12,13,14,15,16,17,18]. The magnetism in uranium ions comes from $5f$ electrons, which in most uranium compounds are in a crossover region between localized and itinerant behavior. The strength of the localization depends strongly on a subtle balance between electronic correlations, crystal field and spin-orbit coupling. It is often rather difficult to decide, on the basis of the experimental data, between Kondo behavior of well localized $5f^2$ configuration and mixed-valence behavior. If the majority of uranium ions are in $5f^2$ configuration, in which two f-electrons are bound into spin $S = 1$, these uranium compounds can be studied in the framework of the Underscreened Kondo lattice (UKL) model. This model considers a periodic lattice of magnetic atoms with localized $S = 1$ spins interacting with conduction electrons via an intra-site antiferromagnetic Kondo coupling and among them via an inter-site ferromagnetic interaction.

The overall physics of heavy fermion compounds can be well described by a generalized Doniach phase diagram. The original Doniach phase diagram has been proposed to describe the experimentally obtained phase diagram of some cerium compounds[5]. In this phase diagram a quantum phase transition between a magnetically ordered one phase and a non-magnetic Kondo phase is observed, i.e. Kondo screening and RKKY coupling are in competition. The Doniach diagram was later extended[4,5] to include the short range magnetic correlations that survive inside the Kondo phase.

The competition between spin glass, magnetism, either ferro- or antiferromagnetism, and Kondo effect can be also analyzed in the framework of a Doniach phase diagram for the disordered Kondo lattice model. In the case of antiferromagnetism, the obtained phase diagram shows the sequence of spin glass, antiferromagnetic order and Kondo state for increasing Kondo coupling J_K[19,21]. The experimental phase diagram of $Ce_2Au_{1-x}Co_x\ Si_3$[9] can be addressed if the J_K coupling is associated with the concentration of cobalt. In the case of ferromagnetism, the phase diagram displays several phase transitions among a ferromagnetically ordered region, a spin glass one, a mixed phase and a pure Kondo state[22]. This phase diagram could be used to partially address the experimental data for $CeNi_{1-x}Cu_x$[6,7].

An analog of the Doniach phase diagram can be derived for the UKL model, and it appears to be significantly different from the original one for the Kondo lattice model with localized spins $S = 1/2$. When decreasing the temperature, the UKL model exhibits two continuous transitions (more precisely, sharp crossovers): the first one, at $T = T_K$, to a non-magnetic Kondo state, and the second one, at $T = T_C$, to a ferromagnetic state that coexists with the Kondo state. At $T = 0$, for a strong Kondo coupling, J_K, ferromagnetism and Kondo effect coexist. The obtained phase diagram has regions of Kondo-ferromagnetic coexistence, non-magnetic Kondo behavior and pure ferromagnetism. This phase diagram can be considered as a new ferromagnetic Doniach diagram for the UKL model.

In this review we analyze how the interplay between Kondo effect, magnetic order, antiferromagnetic or ferromagnetic, and spin glass behavior modify the Doniach phase diagram for the Kondo lattice. We argue that the Doniach phase diagram is a useful theoretical tool for studying such competition, and, at the same time, provides a convenient playground for experimentalists to study the stability of magnetic order in concentrated rare earth and actinide based compounds.

II. S=1/2 KONDO LATTICE MODEL: CERIUM COMPOUNDS.

In a large number of cerium compounds, Ce ions are in the localized $4f^1$ configuration corresponding to spin $S = 1/2$. The localized spin couples antiferromagnetically, via an on-site exchange interaction, J_K, to the conduction electron spin density. At very low temperatures the localized spin $S = 1/2$ is completely screened by the conduction electrons, leading to the formation of coherent Kondo spin-singlet state. Besides, the local coupling between $f-$spins and conduction electrons may give rise to a magnetic order through the RKKY interaction.

Most cerium compounds exhibit antiferromagnetic correlations between Ce ions, and many of them order antiferromagnetically, with rather low Néel temperatures, about 10 K. At low temperatures there is a strong competition between the Kondo effect and antiferromagnetic order. This competition has been firstly described by the well-known Doniach diagram. Accordingly, cerium compounds are separated into those which do not order magnetically and have a very large electronic specific heat constant γ (like $CeAl_3$), and those which order magnetically and present a relatively smaller heavy fermion behavior at low temperatures (like $CeAl_2$). Detailed reviews and references can be found in refs.[3,4]. On the other hand, there are very few compounds which exhibit both a Kondo character and a ferromagnetic order (generally with a low Curie temperature). For example, a recently studied compound is CeRuPO which order ferromagnetically at $T_C = 15K$ and presents a relatively weak Kondo effect[23].

The Hamiltonian of the KL model can be written as:

$$\begin{aligned} H = &\sum\nolimits_{k\sigma} \epsilon_k n^c_{k\sigma} + \sum\nolimits_{i\sigma} \epsilon_0 n^f_{i\sigma} + J_K \sum\nolimits_i \mathbf{S}_i \sigma_i \\ &+ \tfrac{1}{2} J_H \sum\nolimits_{ij} \mathbf{S}_i \mathbf{S}_j \end{aligned} \tag{1}$$

where the first term represents the conduction band with dispersion ϵ_k, width $2D$ and constant density of states $1/2D$; ϵ_0 is a Lagrange multiplier which is fixed by a constraint for the total number of f-electrons per site, $n_f = 1$, and can be interpreted as a fictitious chemical potential for f-fermions. The third term is the on-site antiferromagnetic Kondo coupling, $J_K > 0$, between localized $\mathbf{S_i} = 1/2$ and conduction electron's $\sigma_i = 1/2$ spins. Finally, the last term is the antiferromagnetic inter-site

interaction, $J_H > 0$, between localized f-magnetic moments.

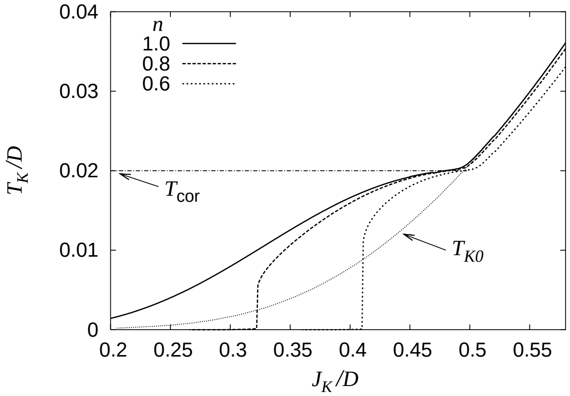

FIG. 1: The non-magnetic region of the Doniach diagram for Ce compounds: Plot of the Kondo temperature T_K as a function of J_K for $J_H/D = 0.04$ and representative values of the conduction band filling, n. It is also showed the correlation temperature $T_{\rm cor}$, and the single-impurity Kondo temperature $T_{\rm K0}$. Reproduced from ref.[5]

This Hamiltonian has been studied within a generalized mean field scheme where the relevant order parameter related to the Kondo effect is $\lambda_\sigma = \langle c^+_{i\sigma} f_{i\sigma}\rangle$. A finite value of λ corresponds to a Kondo state (for a full discussion of the limits and validity od this approximation see references[4,5,24]). Within this mean field treatment a non-exponential Kondo temperature can be obtained, and also a correlation critical temperature for the short-range magnetic correlations near the quantum critical point. In Fig. 1 we show the results of the Kondo temperature as a function of J_K in the non-magnetic region. It is evident that for intermediate values of J_K the Kondo temperature clearly deviates from the one-impurity exponential behavior, and also a short range magnetic correlation between neighboring spins is present, which is characterized by a correlation temperature T_{cor}. For high values of the interaction J_K the Kondo temperature becomes independent of the electron concentration and almost equal to the one-impurity value. These results account for the behavior of T_K in some cerium compounds where a deviation from the exponential law has been observed[4,5]. We remark that in the KL model there is no coexistence between the Kondo region and the magnetic ordered phase. If one includes possible magnetic solution, a narrow region of coexistence is obtained, but either the Kondo or the magnetic solution is the minimum free energy one, it was discussed in ref.[25]). Because of this reason the magnetic region, corresponding to low values of J_K, is not depicted in Fig. 1.

III. THE SPIN GLASS AND MAGNETISM IN KONDO LATTICE.

The second theoretical study we present here concerns the phase diagram of the disordered KL the model. The DKL model is an extension of KL model with a random intersite interaction between the localized spins. The Hamiltonian describing this situation is given by:

$$\begin{aligned} H = &\textstyle\sum_{k\sigma} \epsilon_k n^c_{k\sigma} + \sum_{i\sigma} \epsilon_0 n^f_{i\sigma} + H_{SG} \\ &+ J_K \textstyle\sum_i (S^+_i s^-_i + h.c.) \end{aligned} \qquad (2)$$

where the term $H_{SG} = \sum_{i,j} J_{ij} S^z_i S^z_j$ is given by a quantum Ising interaction between the z-components of the localized spins, and it describes a spin glass character. There are several ways how one can introduce randomness into exchange integrals J_{ij}. First, the exchange integrals can be considered as random variables with a Gaussian distribution with zero average, like in the Sherrington-Kirkpatrick model [26]. In this case the obtained phase diagram shows first a spin glass phase and then a Kondo phase with increasing J_K [27]. Important improvements can be achieved when a non zero average value of the Gaussian distribution is considered. Then a more complex phase diagram with a ferromagnetic [20] or an antiferromagnetic [19] phase occurs at low temperatures for smaller J_K values. This approach allows also to derive a quantum critical point for the magnetic phase [21,28].

Here we discuss some recent theoretical developments which allows to better understand the spin glass and Kondo state coexistence which have been experimentally observed in several cerium [6,7] and uranium [10,11] alloys. This approach is a generalization of the Mattis model [29], and it represents an interpolation between a ferromagnet and a highly disordered spin glass (Detailed calculations can be found in Ref. [22]). The coupling between spins on different sites, i and j, is given by:

$$J_{ij} = \frac{1}{N}\sum_\mu J\xi^\mu_i \xi^\mu_j, \qquad (3)$$

where the $\xi^\mu_i = \pm 1$ ($\mu = 1, 2, ..., p$; $i = 1, 2, ..., N$) are independent randomly distributed variables. For the classical Ising model, if $\mu = 1$, the original Mattis model [29] is recovered. However, if p=N and the N^2 random variables ξ^μ_i have zero mean and unitary variance, , J_{ij} tends to a Gaussian variable in the limit of N large, as in the Sherrington-Kirkpatrick model[26]. Therefore, one can consider this model as an interpolation between ferromagnetism and highly disordered spin glass.

The critical parameter of the model is the ratio $a = p/N$, which describes the degree of the frustration induced by disorder. For large values $a > a_c$, where a_c is a particular value, the frustration is dominant leading to a spin glass behavior, characterized by a certain temperature T_f. At small value of $a < a_c$, this parameter gives

an estimation of the relative importance of the ferromagnetic and spin glass phases for small J_K values, because for large J_K values, the Kondo phase is always present. In fact, for low values of both a and J_K a complex behavior can appear. Therefore, in the presence of disorder, the parameter a together with J_K/J constitute the parameter space of the extended Doniach phase diagram of the DKL model.

In Fig. 2 we present the phase diagram for $a = 0.04$. For a relatively small J_K/J ratio and when decreasing the temperature, there is a a spin glass phase, then a coexistence region between ferromagnetism and spin glass and, finally, a ferromagnetic phase.

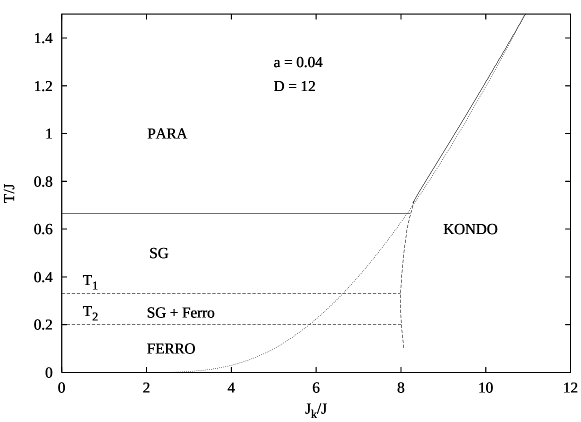

FIG. 2: The Kondo-spin glass-ferromagnetic diagram versus J_K/J for D=12 and a=0.04.

Fig. 2 accounts very well for the experimental phase diagram of $CeCu_{1-x}Ni_x$ disordered alloys, where Kondo, spin glass and magnetically ordered phases are observed. The experimental phase diagram of $CeCu_{1-x}Ni_x$ is in fact very complicate: for example,a very careful experimental study of $CeCu_{0.6}Ni_{0.4}$ yields a percolative transition with decreasing temperature from a cluster-glass state with ferromagnetic correlations to a ferromagnetic clustered state and recent theoretical simulations can reproduce satisfactorily the experimental situation[8]. But the results presented in Fig. 2, and in a more detailed way in Ref.[22], provide a fairly good description of the phase diagram of $CeCu_{1-x}Ni_x$ alloys.

IV. THE UNDERSCREENED KONDO LATTICE APPLIED TO URANIUM COMPOUNDS

As we already said in the introduction, uranium compounds show very rich behaviors, quite different from cerium compounds, with the presence of numerous coexistence phenomena. Here we discuss mainly the coexistence of magnetic order with Kondo effect, which can be well described in the framework of a extended ferromagnetic Doniach diagram. Let us briefly describe the experimental situation. The first experimental evidence of the coexistence between Kondo behavior and ferromagnetic order in the dense Kondo compound UTe has been obtained long time ago[12]. More recently, this coexistence has been observed in $UCu_{0.9}Sb_2$ [14] and $UCo_{0.5}Sb_2$ [15,16]. All these systems undergo a ferromagnetic ordering at the relatively high Curie temperatures of $T_C = 102$K (UTe), $T_C = 113$K ($UCu_{0.9}Sb_2$) and $T_C = 64.5$K ($UCo_{0.5}Sb_2$). Above the ordering temperatures, i.e. in the expected paramagnetic region, these materials exhibit a Kondo-like logarithmic decrease of the electrical resistivity, indicating a Kondo behavior. This logarithmic variation extends down to the ferromagnetic Curie temperature, T_C, suggesting that the Kondo behavior survives inside the ferromagnetic phase, implying that the ferromagnetic order and the Kondo behavior do coexist. This coexistence, together with the large Curie temperatures, are clearly novel features that cannot be explained by the standard KL model.

This coexistence can be qualitatively understood by studying the underscreened Kondo lattice model. Detailed results can be found in refs.[24,30]. The UKL model describes the situation when the localized spins are larger than 1/2, and therefore, they cannot be completely screened by the conduction electrons at $T = 0$. The UKL model can be applied to uranium compounds if the majority of uranium ions are in $5f^2$ configuration, in which two f-electrons are bound into spin $S = 1$. We believe this is an appropriate description of electronic states for those compounds which have magnetic moments close to the free ion values[12,14,15]. Here we will describe a mean field solution of the UKL model, which describes well the physics of the above mentioned uranium compounds.

The Hamiltonian of the UKL model can be formally written as Eq.(1), the one for the normal KL model. The main difference between the KL Hamiltonian (Eq.(1)) and the UKL model is that the localized spins are now $\mathbf{S_i} = 1$. Correspondingly, the constraint for the total number of f-electrons per site is modified, and it is given by $n_f = \sum_\sigma (n_\sigma^{f1} + n_\sigma^{f2}) = 2$, where $f1$ and $f2$ are two degenerate f-orbital states. Also, the last term of the Hamiltonian now represents a ferromagnetic inter-site interaction between localized f-spins, then for the UKL Hamiltonian $J_H < 0$.

We restrict our considerations to a mean-field (MF) treatment of the Hamiltonian and introduce four MF parameters $\lambda_\sigma = \langle \sum_\alpha c_{i\sigma}^+ f_{i\sigma}^\alpha \rangle$, $M = \frac{1}{2}\langle n_{i\uparrow}^f - n_{i\downarrow}^f \rangle$ and $m = \frac{1}{2}\langle n_{i\uparrow}^c - n_{i\downarrow}^c \rangle$, where $\langle ... \rangle$ denotes the thermal average.

Non-zero values of M and m describe a magnetic phase with non-zero total magnetization, while non-zero values of λ_σ describe the Kondo effect and the formation of the heavy-fermion state, as in the case of the standard KL. For large values of J_K, one obtains a Kondo-ferromagnetic state below the Curie temperature T_c, then a Kondo state between T_c and the Kondo temperature T_K and finally a paramagnetic state above T_K. Then, the critical temperatures, T_c and T_K, have been derived as a function of J_K. We would like to stress that T_K

increases abruptly above a critical value J_K^c, while T_c is always non zero but increases slowly above J_K^c and then remains at a value smaller than T_K. This ferromagnetic-Kondo diagram is shown in Fig. 3 and is completely different from the Doniach diagram derived for cerium compounds. Thus, the UKL model provides the first explanation of the behavior of ferromagnetic Kondo uranium compounds, describing the coexistence of Kondo behavior and ferromagnetic order.

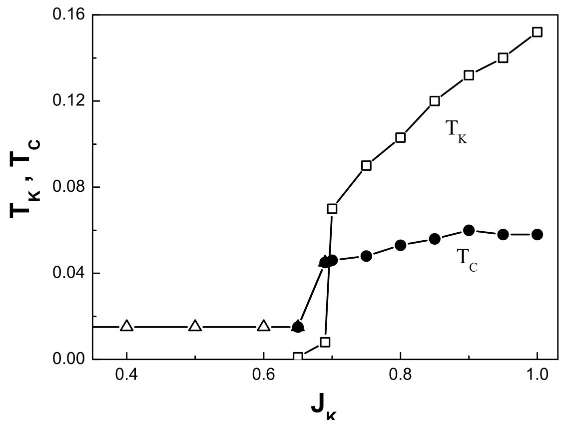

FIG. 3: The ferromagnetic Doniach diagram: Plot of the Curie temperature T_C and the Kondo temperature T_K versus J_K for $J_H = -0.01$ and $n_c = 0.8$. Reproduced from Ref.[24]

Besides its applicability to the physics of ferromagnetic uranium compounds, the UKL model is an interesting problem on its own, but yet it has attracted a little attention. Possible extensions of this model would be very interesting, in order to describe the coexistence between the ferromagnetic order and Non-Fermi-Liquid behavior, as observed in $URu_{2-x}Re_xSi_2$ compounds[31] or the decrease of T_c versus J_K down to a eventual quantum critical point.

V. CONCLUSIONS

We have analyzed how the interplay between Kondo effect, magnetic order, (antiferromagnetic or ferromagnetic), and spin glass behavior modify the Doniach phase diagram for the Kondo lattice. We have discussed in detail the cases of a) cerium compounds where the Kondo effect coexists with short range magnetic correlations but competes with long range magnetic order, b) disordered cerium alloys of the type $CeCu_xNi_{1-x}$, where the obtained Doniach diagram exhibits magnetic ordered, spin-glass and Kondo phases and c) uranium compounds, like UTe, which are well described by the UKL model, that can account for the observed coexistence between ferromagnetic order and Kondo behavior observed in those uranium compounds.

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